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(54) Selective delivery and retention of aldehyde and nicotine by-product from cigarette smoke.

(57) A cigarette filter element containing compacted fibrous polyolefinic substrate material the the surface of which is coated with about 5-50% by weight of a modifying composition containing polyethyleneimine and formic, propionic, butyric, lactic, benzoic or acetic acid as a buffer, and a process for making the modified filter element by treating the substrate material with the modifying composition buffered to a pH in the range of 8 to 9.5.

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SELECTIVE DELIVERY AND RETENTION OF ALDEHYDE AND NICOTINE BY-PRODUCT FROM CIGARETTE SMOKE

This invention relates to fibrous cigarette filter elements containing compacted synthetic thermoplastic substrate material treated with a filter-modifying composition.

The term "substrate" refers to any conventionally used fibrous filtering media used for insertion into the garniture of a conventional apparatus for forming compacted filter rods, including fibers in the form of opened fiber tow, a ribbon of nonwoven material, sliver, or fibrillated film.

Tobacco-smoke filters comprising a compacted synthetic fiber components, particularly polyolefins such as polypropylene, are well known and have been used in the filter-tip part of cigarettes for several years. Such synthetic fiber components are desirable because they can be easily drawn to a small denier and can provide high filter efficiency combined with the strength needed for crimping and the tension resulting from high speed production.

Synthetic fiber components, particularly polyolefins such as polypropylene, however also have the disadvantage that they are normally hydrophobic and tend to be chemically inert, while a majority of the additives used to enhance filtering efficiency tend to be hydrophilic and difficult to retain within filters comprising hydrophobic synthetic fiber. They may also present difficulties in achieving desirable combinations of physical properties such as general filtering efficiency, dimensional stability, hardness, and satisfactory "draw" (pressure drop across the filter element), which depends substantially upon fiber denier, filter length and density, and length, while achieving a high degree of selective filtration of cigarette smoke components, in particular the ability to conduct flavororants such as nicotine efficiently while efficiently limiting the passage of less desirable smoke by-products.

While various synthetic fibers and fiber mixtures have been tried and evaluated as filter components, a substantial number of contemporary cigarette filter producers continue to use old technology and substrate combinations because of cost and handling advantages. For example, cellulose acetate tow using a variety of additives can be processed into cuttable filter rods using traditional filter rod-making apparatus, and there is resistance to innovations that require equipment changes.

U.S. Patent 4,266,561 discloses chemisorptive compositions, incorporated in tobacco smoke filters, that are prepared by treating aluminium oxide particles with about 5% of polyethyleneimine and coating the particles with 0.5% to 5% of an aqueous buffering solution of acetic acid and zinc acetate to produce a pH of 6.0 to 7.6 prior to drying. The particles are then dispersed throughout the fibers of the filter, and are said to remove hydrogen sulphide, hydrogen cyanide, and acetaldehyde from the smoke. The use of particulate carriers for similar additives is common in the art (e.g., see U.S. Patents 3,716,063, Re 28,858 and 3,428,056) and unbuffered polyethyleneimine on a non-olefinic substrate is disclosed also in U.S. Patent 3,340,839.

There is a need for a less laborious and more efficient process for the high-speed production of fibrous cigarette filter elements containing compacted polyolefinic substrate material, that facilitates multi-selective filtration abilities, such as removal of aldehyde by-products while improving the delivery of flavors such as nicotine, without causing undesirable discoloration of the filter or adversely changing the fiber content and hardness characteristics, and without reducing the ability to use conventional mono-, or bi-component configurations of fibers in conventional filter rod-making apparatus.

According to the invention, a cigarette filter element containing compacted fibrous polyolefinic substrate material in which is incorporated polyethyleneimine and a carboxylic acid is characterized in that the surface of the substrate material is coated with about 5 to 50% (preferably 5 to 25%) by weight of a modifying composition containing polyethyleneimine ("PEI") and formic, propionic, butyric, lactic, benzoic or acetic acid, based on the total weight of dry substrate material

Also according to the invention, a process for modifying the filtration characteristics of a cigarette filter element containing compacted thermoplastic substrate fibers is characterized in that a filter modifying solution containing polyethyleneimine ("PEI") and formic, propionic, butyric, lactic, benzoic or acetic acid and having a pH within the range of about 8 to about pH 9.5 is directly applied to the surfaces of the substrate and the fibers are dried under humidity conditions used for filter storage for instance, 55-65% relative humidity.

The application of the modifiers and additives to the substrate can be carried out, for instance, by dipping, spraying or by drawing a solution through a formed filter rod or element, using a partial vacuum.

Modifier compositions having a pH above 8 are particularly since they achieve the above advantages without the possibility of causing discoloration of the treated substrate.

Cigarette filter elements of the present invention comprise compressed polyolefinic substrate material,

containing polyolefin fibers, alone or in combination with cellulose acetate fibers, using regular "plug wrap" such as paper having a weight within a range of about 25-90 g/m² or higher.

Conventional surfactant material amounting to about 0.1%-10% and preferably 0.5%-10% by weight of one or more of polyoxyalkylene derivatives of a sorbitan fatty acid ester, a fatty acid monoesters of a polyhydroxy-alcohol, or fatty acid diesters of a polyhydroxy alcohol may also be incorporated according to known practice.

In addition to the modifier compositions and surfactants there may be included conventional additives, including aqueous solutions, suspensions or dispersions of humectants such as polyhydric alcohols such as glycerols and glycols, and flavors and perfumes.

The term "substrate" as above used, includes generally all the fibrous materials suitable for feeding into conventional apparatus for making filter rods. Particularly if not all of the substrate in the filter element is to be used as a carrier surface for the filter modifier composition, a nonwoven fabric of the same or different fiber composition and denier may be included in the substrate.

The substrate such may comprise about 5%-100% by weight of a polyolefin, including mono-, or bi-component fiber of side-by-side and sheath/core types, and may consist of conventional webs or tows having filaments of homogeneous or mixed denier, or conventional combinations of two or more kinds of fibers such as polypropylene/polyethylene, polypropylene/polyvinylidene chloride, polypropylene/cellulose acetate, polypropylene/rayon, polypropylene/nylon, cellulose acetate/polyethylene, polypropylene/paper, polypropylene/polystyrene/polyethylene, and the like, in preferred ratios of about 10%-90% 90%-10% or 10%-90%.45%-5%/45%-5% based on substrate weight, as well known from, for instance, in USP 3,393,685.

Generally speaking suitable nonwoven material that falls within a conventional weight range of about 10-50 grams per m², and a ribbon width of about 10cm-30.5cm of such substrate material will permit successful passage through the garniture of a conventional filter rod-making apparatus operating at production speeds. Fibrillated film can of course be conventionally employed as a substrate, alone or in combination with other substrate components.

A conventional filter rod-making apparatus suitable for present purposes comprises a tow trumpet, garniture, shaping means, wrapping means, and cutting means in accordance with well-known practice. Conventional modifications can be made to permit in-situ or prior spraying, dipping, printing, or vacuum draw for introducing the modifying compositions.

The inclusion of an additional low melting fiber such as polyethylene, combined with other fiber as garni- ture feed is also found useful for obtaining filter elements of various bonding and adsorption properties.

In order to maintain precise control over application of the modifying compositions and additives, the moisture content of the substrate should be carefully controlled as above noted before conversion into a filter element.

The filter elements and applied components are preferably isolated from direct contact with the lips by applying the active components onto a substrate material that is conventionally sandwiched within two or more untreated nonwoven fabrics of lesser permeability.

Where a continuous fiber tow is used as a substrate component, preparation of the tow is conveniently carried out in the usual way by drawing the fiber from one or more creels through a conventional fluid bulking or texturing jet, and then handled as noted above.

Substrates that are employed in the above manner can usefully comprise a variety of synthetic filaments, including polyesters, polyamides, acrylics, as well as polypropylene and the like. Due to its relatively low density, compared to other synthetic fiber-forming material and its excellent spin properties, combinations of filament-forming copolymers of propylene with ethylene or other lower olefins monomers are particularly preferred as tow, nonwoven ribbon (of monofilament or bicomponent fiber or fiber webs) and fibrillated film material.

The bulk denier of a tow for carrying out the present invention can conventionally fall between about 2,000 and 10,000, and this substrate can be supplied as a crimped fiber from a single creel or bale, or as a composite of several creels or bales combined and passed through a fluid jet simultaneously.

The invention is further illustrated by the following Examples:

Example 1

(A) Baled 4.5 dpf "y" cross section polypropylene fiber obtained from melt spun isotactic polypropylene having a flow rate of 35.2gm/10 minutes, is broken, opened, carded, crimped and pulled to form

a thin tow ribbon about 12-14 inches in width. The ribbon is drawn, without further treatment, through the garniture of a conventional filter rod-forming apparatus, here identified as model PM-2, obtained from Molins Ltd. of Great Britain, and compressed to form filter plugs which are wrapped with BXT-100 polypropylene film to form 108 mm test filter rods. The rods are then cut into 27 mm lengths of essentially equal weight, and draw; all tested filter elements have a resistance to draw (RTD) within the range of 111-136 mm Wg (water gauge). Some of test filter rods are taped onto R. J. Reynolds' Camel Light tobacco plugs, and stored for 48 hours in a humidity cabinet at 55%-65% relative humidity at 22° C according to CORTESTA Standard Method #10; the remaining unattached filters are air dried at 70° C. and stored in the humidity cabinet at 55%-65% relative humidity for 48 hours prior to testing.

(B) Total particulate/nicotine determinations are obtained by smoking ten filter-tobacco plug test cigarettes as described in Ex 1A, at a rate of one 35 cc puff/minute down to 35 mm lengths, using a Borgwaldt smoking machine, Model # RM-1/G. The particulate matter in the resulting filtered smoke is trapped on a preweighed Cambridge filter pad, and the pad reweighed after smoking, to determine the amount of particulate matter (TPM) which is passed through each treated or untreated cigarette filter. The Cambridge pad is then soaked overnight in anhydrous isopropyl alcohol, and the extract conventionally tested for nicotine and water content using a GC (gas chromatograph) autosampler, Hewlett Packard Model HP5890.

(C) Aldehyde determinations are also run on a 10 cigarette sample basis using filter elements of Example 1A by directing a measured volume of filtered cigarette smoke into a collection bottle containing a saturated 2.2N HCl solution of 2,4-dinitrophenylhydrazine (DNPH) and 25 ml methylene chloride; the bottle is shaken for 2 hours, and the phases allowed to separate. Aliquot samples of the methylene chloride phase are then removed by syringe for conventional (HPLC) aldehyde analyses.

Test results of Example 1B and 1C are individually averaged and reported in Table I below as S-1 through S-3 and as corresponding C(Control) numbers C-1 through C-3.

TABLE 1

SAMPLE	TREATMENT*5 ACTIVE COMPONENT	% ACETALDEHYDE REMOVED	% ACROLEIN REMOVED	% FORMALDEHYDE REMOVED	NIC*6 TPM (%) PASSED	FILTER*7 COLOR
S-1*9	2% PEI + acetic acid (pH 4.5)	32	27	79	7.2	SD
C-1	Control	--	--	--	5.5	---
S-2*9	5% PEI + formic acid (pH 8)	38	28	56	8.2	D
C-2	Control	--	--	--	6.3	---
S-3*8	5% PEI + formic acid (pH 8)	27	26	71	9.3	D
	C-3	Control	--	--	--	4.5
	---					

*5 Average/10 test Cigarettes using treated filter elements secured to Reynolds Light tobacco plug.

*6 Collected in 2,2NHCl containing DNPH and methylene chloride. Conventional HPLC analysis run on methylene chloride aliqnot.

*7 D = Slight discoloration/filter.

SD = Significant discoloration/filter

*8 Applied filter modifier onto precrimped polypropylene open tow by using a double-spray, then dried at 55%-65% relative humidity.

*9 Filter modifier injected directly into filter and the filter dried at 60° C. for 24 hours then stored for 48 hours at 55%-65% relative humidity.

Example 2

A. Test filter elements as described in Example 1 are individually injected with one (1) ml samples of 5% solution of PEI adjusted to a pH of 2, 4, 6, 8, 9 and 10 by dilution with concentrated acetic acid (S-4 through S-8) or formic acid S-9 through S-14; the treated filter elements are then dried and stored under controlled humidity as described in Example 1.

B. Unattached filter elements described in Example 2A are endwise secured by air-tight connection to Tygon tubes on one side through a check value to a gas bag containing a 5 ppm acetaldehyde/air mixture, and on the opposite side to a Borgwaldt smoking machine, Model # RM-1/G, adjusted for five two (2) second 35 cc puffs over a ten (10) minute period. The filtered test gas is collected in a gas sampling loop and analyzed at 150°C. using a Varian 3300 model gas chromatographer equipped with a flame ionization detector to determine the through concentration.

Test results are tabulated, using a Varian Model 4290 integrator and reported in Table II as S-4 through S-14.

TABLE II

Filter Modifier	Sample	pH	% Acetaldehyde Removed	Acetaldehyde Passed (ppm)
HAc/PEI	S-4	4	50	2.5
---	Control	--	--	5.0
HAc/PEI	S-5	6	50	2.4+
---	Control	--	--	5.0
HAc/PEI	S-6	8	45	3.7+
---	Control	--	--	5.0
HAc/PEI	S-7	9	40	3.0
---	Control	--	--	5.0
HAc/PEI	S-8	10	26	3.7
---	Control	--	--	5.0
Formic/PEI	S-9	2	23	3.8+
---	Control	--	--	5.0
Formic/PEI	S-10	4	49	2.5+
---	Control	--	--	5.0
Formic/PEI	S-11	6	71	1.4+
---	Control	--	--	5.0
Formic/PEI	S-12	8	76	1.2
---	Control	--	--	5.0
Formic/PEI	S-13	9	51	2.4+
---	Control	--	--	5.0
Formic/PEI	S-14	10	24	3.8
---	Control	--	--	5.0

Example 3

Example 2 is repeated but using filter elements injected respectively with 1 ml of 5% PEI modified by formic, propionic, butyric, benzoic, lactic, or acetic acids to pH values of 8 or 6. The dried and stored filter elements are processed as described in Example 2A and secured to a test gas bag (5 ppm acetaldehyde/air) and a Borgwaldt smoking machine as described in Example 2B. Test results are collected as before and reported in Table III.

TABLE III

SAMPLE	TREATMENT[8] ACTIVE COMPONENT	% Acetaldehyde Removed	Acetaldehyde Passed (ppm)
S-1	5% PEI + formic acid (pH 8)	80	1.0
Control	-----	--	5.0
S-2	5% PEI + propionic acid (pH 6)	65	1.7 +
Control	-----	--	5.0
S-3	5% PEI + butyric acid (pH 6)	53	2.3 +
Control	-----	--	5.0
S-4	5% PEI + benzoic acid (pH6)	19	4.0 +
Control	-----	--	5.0
S-5	5% PEI + lactic acid (pH 6)	44	2.8
Control	-----	--	5.0
S-6	5% PEI + acetic acid (pH 8)	50	2.5
Control	-----	--	5.0

Claims

1. A cigarette filter element containing compacted fibrous polyolefinic substrate material is characterized in that the substrate material is coated with about 5-50% by weight of a modifier composition containing polyethyleneimine ("PEI") and formic, propionic, butyric, lactic, benzoic or acetic acid as a buffer.

2. A process for modifying the filtration characteristics of a cigarette filter element containing compacted thermoplastic substrate fibers is characterized in that a treating solution containing polyethyleneimine ("PEI") and formic, propionic, butyric, lactic, benzoic or acetic acid and having a pH within the range of about 8 to about pH 9.5 is directly applied to the surfaces of the fibers and the filters are dried.

3. A process as claimed in claim 2, further characterized in that 5-25% by weight of the modifier composition, based on total weight of dry filter substrate, is applied.

4. A process as claimed in claim 2 or 3, further characterized in that the filters are dried under 55%-65% relative humidity at 22° C.

5. A process as claimed in claim 2, 3 or 4, further characterized in that the substrate material comprises about 5%-100% by weight of polypropylene.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 12 3501

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
D,Y	US-E- 28 858 (LITZINGER) * Comparative example II; column 2, line 47 - column 3, line 36 *	1,5	A 24 D 3/10 A 24 D 3/14
D,A	---	2,3	
D,Y	US-A-3 340 879 (HORSEWELL et al.) * Column 7, lines 41-57; claims 1,2,8-10; examples 1,13,14 *	1,5	
D,A	---	4	

TECHNICAL FIELDS SEARCHED (Int. Cl.5)

A 24 D

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	23-03-1990	LEPRETRE F.G.M.J.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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